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Health Effects Research Lab., Research Triangle Park, N.C.

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Environmental Health Effects Research Series

WORKSHOP ON HEALTH EFFECTS OF TRANSPORTATION-RELATED POLLUTANTS

Health Effects Research Laboratory
Office of Research and Development
U.S. Environmental Protection Agency
Research Triangle Park, North Carolina 27711

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The Department of Transportation, recognizing the need for further health effects research data in assessing the environmental implication of its policies over the next several years, called upon the Environmental Protection Agency's health effects group to bring together key experts in the field to address both DOT's and EPA's research needs and to evaluate resources available.

This report is a brief summary of the presentations and conclusions of the DOT-EPA sponsored Workshop on Health Effects of Transportation-Related Pollutants held at the Sheraton-Crabtree Motor Inn, Raleigh, North Carolina, on June 15-16, 1977. This report is, in part, interpretive and intended to supply a synopsis and not a verbatum transcript of the proceedings.

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WORKSHOP ON HEALTH EFFECTS OF TRANSPORTATION-RELATED POLLUTANTS

Sheraton-Crabtree Motor Inn

Raleigh, North Carolina

June 15-16, 1977

Sponsored by

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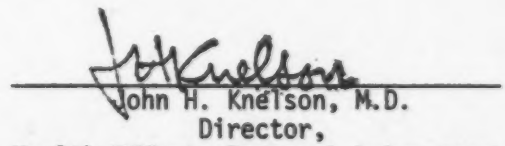
FOREWORD

The many benefits of our modern, developing, industrial society are accompanied by certain hazards. Careful assessment of the relative risk of existing and new man-made environmental hazards is necessary for the establishment of sound regulatory policy. These regulations serve to enhance the quality of our environment in order to promote the public health and welfare and the productive capacity of our Nation's population.

The Health Effects Research Laboratory, Research Triangle Park, conducts a coordinated environmental health research program in toxicology, epidemiology, and clinical studies using human volunteer subjects. These studies address problems in air pollution, non-ionizing radiation, environmental carcinogenesis and the toxicology of pesticides as well as other chemical pollutants. The Laboratory develops and revises air quality criteria documents on pollutants for which national ambient air quality standards exist or are proposed, provides the data for registration of new pesticides or proposed suspension of those already in use, conducts research on hazardous and toxic materials, and is preparing the health basis for non-ionizing radiation standards. Direct support to the regulatory function of the Agency is provided in the form of expert testimony and preparation of affidavits as well as expert advice to the Administrator to assure the adequacy of health care and surveillance of persons having suffered imminent and substantial endangerment of their health.

The Department of Transportation, recognizing the need for further health effects research data in assessing the environmental implication of its policies over the next several years, called upon the Environmental Protection Agency's health effects group to bring together key experts in the field to address both DOT's and EPA's research needs and to evaluate resources available. Both the conclusions regarding priorities and the supporting rationale are included in this report.



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ABSTRACT

This report is a brief summary of the presentations and conclusions of the DOT-EPA sponsored Workshop on Health Effects of Transportation-Related Pollutants held at the Sheraton-Crabtree Motor Inn, Raleigh, North Carolina, on June 15-16, 1977. This report is, in part, interpretive and intended to supply a synopsis and not a verbatum transcript of the proceedings.

ACKNOWLEDGMENT

The initiative for this conference was provided by Dr. Richard Strombotne, Director, Office of Energy and Environment, Office of the Secretary, Department of Transportation. Dr. Strombotne's recognition of the need for such a session was translated into action by Mr. Samuel Coroniti, of his staff, who made the contacts with appropriate EPA staff and thereafter coordinated the joint effort of bringing together key experts to address the objectives of EPA and DOT.

In addition, we would like to express our appreciation to Dr. Daniel B. Menzel, Duke University Medical Center, for serving as reporter at the workshop and for the numerous hours spent in preparation of this document.

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I. EXECUTIVE SUMMARY OF CONFERENCE OBJECTIVES AND CONCLUSIONS

A. General Issues: The following general issues were discussed and recommended by the workshop.

1. The complexity of the transportation-associated air pollutants is such that a complete evaluation of the presently recognized health hazards to man is not achievable in less than 10 years. Short-term goals which have importance and relevance to present health hazards, however, can be achieved.

2. Present facilities and personnel for the study of air pollutants are inadequate to evaluate the presently known pollutants. Development of a stable cadre of scientists and physical facilities to meet the added objectives of the DOT program are recommended.

3. Research strategies presently being applied to the general study of air pollutants may be applicable to the gap areas in transportation-related air pollutants identified by this workshop. A detailed study of most effective strategies for future studies is needed. Formation of an expert multidisciplinary advisory study group to develop such strategies for DOT is needed and is recommended.

Examples of methods and strategies of research identified by this workshop as applicable to the DOT program are listed below. It is the opinion of the workshop participants that there are insufficient data to warrant more than preliminary human experimentation research without detailed animal studies. Medical-legal consideration prevents the exposure of volunteers to many of the pollutants identified by the group without first collecting toxicological data in animals. Carcinogenesis is a potential problem area which can only be conducted in experimental animals. Epidemiological studies are needed but are difficult to define since sensitive indicators of biological damage which can be applied to man are only now being developed in animal models. Further constraints on epidemiological studies are the time and complexity of design and collection of appropriate monitoring data. None of these considerations, however, should be taken to infer that human experimentation or epidemiological data are considered inappropriate or of low priority; rather, these studies are considered urgent, first priority projects whenever possible.

Specific methods of procedure discussed and considered to be applicable strategies for implementing research in the areas identified are as follows:

1. Epidemiological studies directed to specific transportation-associated pollutants, including those segments of the population likely to be at greatest risk either geographically, medically, or physiologically;

2. Human clinical studies;

3. Infectivity mortality studies in small animals with both bacterial and viral pathogens;

4. Perfused isolated animal lungs and human lung fragments, as means of studying metabolism, physiology, and pharmacology;

5. Chronic animal exposures followed with conventional and innovative morphological measurements;

6. Isolated cell and organ culture systems;

7. Bacterial and microbial cell systems for mutagenesis and carcinogenesis testing;

8. Whole animal carcinogenesis testing;

9. Teratogenesis testing in whole animals and some limited organ culture systems;

10. Immunobiological systems, especially those systems dealing with cellular mechanisms of immunity;

11. Neurobiological and behavioral evaluation; and

12. Hormonal alterations that might lead to pathophysiology.

Application of these methods of procedure to the specific objectives listed below will depend upon the nature of the problem and is deserving of detailed study and recommendation by the expert interdisciplinary advisory group.

B. Specific Objectives Recommended for the DOT Health Effects Program: Specific areas were identified by the workshop as in need of investigation. These areas were selected on the basis of their immediate relevance to transportation in its broadest sense, including air and ground transportation. Air pollution remains the principle concern and focus relevant to transportation. Since EPA, NIH and others are supporting programs relating to the general problem of air pollution, the areas listed below were considered by the workshop as those areas that were in need of immediate support, but were not included in existing programs.

As the workshop participants came from a broad background, their knowledge of planned as well as on-going research was drawn upon to formulate the objectives listed below. It is the clear intention of the workshop that these objectives were not intended as a comprehensive statement of the general problem of transportation air pollution, but represents the expert opinion of the group as to the minimum program which might be added on to existing programs through DOT support. While these are specialized programs related to transportation in particular, they are still part of the overall problem of air pollution. There is no way in which pollution related to transportation systems can be divorced from the more general problem of air pollution.

Specific objectives were divided into three priorities of immediate, near-term needs, secondary objectives, and long-term objectives. Within these categories no priorities were assigned.

1. Primary Needs:

a. Coordination with Changing Technology: Biological studies must keep pace with the projected changes in power plants used in transportation systems and the consequent changes in exhaust emissions. An example of an important immediate gap is the evaluation of the biological effects of changes from gasoline to diesel power plants. Interaction between transportation technologists and biologists must be assured in all future programs to avoid recurring gaps and to allow sufficient lead time.

b. Human Clinical Studies: Because of the rapidly evolving information on the toxicology of air pollutants, a constant review of the applicability of methods developed from animal and in vitro studies to man is recommended. Human experimentation is recommended in all those areas where it is safe and reasonable.

c. Epidemiological Studies: Support of epidemiological studies is recommended using the same criteria as above for human experimentation. Epidemiological studies should be planned carefully with a detailed evaluation of the pollution monitoring methods prior to implementation. Since epidemiological studies require long time periods for completion, stable support for these studies is recommended. Specific projects for both epidemiological and human experimentation should be evaluated by an expert advisory group.

d. Exhaust Particulates Including Ultrafine Aerosols: Particles arising from auto exhaust and from conversion of exhaust components in the atmosphere is a clearly identified area deserving greater study. An important area is the inhalation toxicology of respirable particles including ultrafine aerosols in general. Knowledge of

the toxicology of ultrafine particles is particularly lacking. The role of particles as carrier of toxicants likewise needs investigation. A critical issue is the sorption of emission products or secondary pollutants on carbon particles of a variety of sizes. These studies are directly related to the biological effects of diesel emissions.

e. Dose-Response Relationships: Dose-response relationships, including the number of exposures, concentration of toxicant, duration of exposure and frequency of exposure, are very important aspects of the toxicity of transportation-associated pollutants. Detailed studies of dose-response relationships are needed for these air pollutants (ozone, NO_2, sulfuric acid and sulfate aerosols, and organic particulates). These data bear directly on estimates of health impacts and modeling of the effects of alternative control strategies.

f. Potentiation of Toxic Effects of Mixtures of Air Pollutants: Since most studies of the toxicity of air pollutants associated with automotive exhaust have been conducted on single air pollutants alone, potentiation of the toxic effects of combinations of air pollutants has not been fully evaluated. Some data indicates a greater toxicity resulting from sulfuric acid and ozone combinations than from these toxicants alone, depending upon the sequence of exposure. Auto exhaust is a mixture of both different chemical compounds and different physical states (particles and gases). Evaluation of the toxicity of gaseous pollutants in the presence of particulate pollutants, especially carbon particles and respirable particles generated as secondary pollutants in the atmosphere, is needed. Specific combinations of pollutants related to transportation that should be evaluated are particulates and oxidants (ozone and nitrogen dioxide), sulfuric acid aerosols and carcinogens, carbon particles and oxidants, and sulfate aerosols and oxidants. Studies on carbon particles as carriers are particularly important to diesel exhaust emission evaluations.

g. Hydrocarbons and Oxygenated Hydrocarbons Emitted into the Atmosphere: Hydrocarbon and oxygenated hydrocarbon species are emitted into the atmosphere from mobile vehicle exhausts. Atmospheric transformation of these compounds may be the most critical step in determining the biological activity of these compounds. Changes in fuel, power plant and emission control devices will effect the chemical composition of these compounds inhaled by man. These compounds may be potential carcinogens or modifiers of the neoplastic process and, therefore, of immediate importance. Detailed data have not appeared identifying these hydrocarbon species (polynuclear aromatic and otherwise) or their oxygenated forms so that biological estimates of their toxicity are difficult. Further study of both the chemical nature and biological action is needed.

h. Toxicology of NO_x Mixtures: Mixtures of pollutants, particularly NO_x, may result in different chemical species depending upon the relative humidity. Nitrous acid may result, for example, from NO_x at high relative humidity to provide concentrations of this compound adequate for interaction with cellular components on inhalation. Nitrous acid may be important in the direct formation of carcinogenic nitrosamines in situ. The presence and reactivity of nitrous acid, nitric acid and similar species depending upon the relative humidity deserves further study and toxicological evaluation.

2. Secondary Needs:

a. Tolerance Mechanisms: Verification and elucidation of the "tolerance" mechanism to air pollutants is a basic concern to the interpretation of human health effects of air pollutants. Studies in man and animals deserve support.

3. Tertiary Needs:

a. Hetero-Organic Compounds: Our knowledge of the complex organic compounds containing S, N, and O originating from exhausts or from atmospheric conversion is insufficient to predict their potential health effects or to provide adequate direction for health effects experiments at present. An evaluation of the biological impact of these compounds should continue with a limited number of compounds selected for biological evaluation based on the likelihood of toxicity from chemical structural similarities with known toxicants.

b. Aircraft and Roadside Deposition: The deposition of particulate materials from aircraft and motor vehicles on foodstuffs, soil, plants, and other means of translocation of these particles to man is also of concern.

c. Particles from Roadway and Vehicle Wear: Inhalation particles arising directly from the roadway are potentially hazardous. The size distribution and the nature of the particles generated from the roadway should be determined since the preliminary data on the composition of the mineral content of some roadways suggests that particles of the respirable range may be highly damaging. While tire wear does not appear to be a health hazard since it does not produce particles of a respirable size, other products of vehicle degradation may deserve investigation.

C. Supporting and Ancillary Research Needs: Despite considerable effort in inhalation toxicology and air pollution research there still exist supporting and ancillary research needs that are presently unmet.

1. Further characterization of emission products and their transformation in the atmosphere is needed.

2. Further development of mathematical models of the deposition of inhaled toxicants in the human and animal lung which can allow a more sound extrapolation from animal exposures to human exposures is needed. A model for human and animal lungs for ozone and other soluble gases has been developed. Improvement of the model for particle deposition is needed. Development of a model for liquid and soluble aerosols is needed.

3. Development of animal models of human diseases which may place the diseased person at greater risk is needed.

II. CONFERENCE SUMMARY

A. Introduction and DOT Objectives (Conference Discussant - Mr. S. Coroniti): The Department of Transportation has an immediate and continuing concern for the health effects of transportation. Legislative mandates to this end are contained in the DOT Act, Clean Air Act, NEP Act, Airport and Airway Development Act, Urban Mass Transportation Act, and the ERDA Act amongst others. Autos and other mobile vehicles contribute a significant proportion of the emissions entering the atmosphere resulting in air pollution. The impact of transportation systems on the degradation of the air quality is a challenge which DOT must meet to fulfill its public obligations in the planning and execution of transportation systems for the nation. Through its review and assistance DOT also has a direct impact upon state and local plans for the development of new transportation modes. State and local governments have turned to DOT for guidance and help in devising strategies designed to reduce the air pollution impact of transportation. To assist in the planning of transportation systems at all levels, DOT is seeking the assessment of this workshop of the areas of concern and the action needed to contend with present day and future health effects. Since the primary route of entry of these pollutants is through inhalation, the workshop objectives have been restricted to air pollutant effects. Other groups will be considering effects on transportation-related pollutants on the remainder of the environment independent of this workshop.

B. Overview of Ongoing Research on Automotive-Related Pollutants:

1. Overall Review (Conference Discussant Dr. D. E. Gardner): The present ongoing research aimed at studying the health effects of automotive-related air pollutants within EPA are set out in Tables 1-3. Both in vivo and in vitro testing methods are being used. A major effort is directed toward the use of the highly sensitive infectivity model of the toxic effect of air pollutants. Both primary and secondary pollutants are being studied. The effects of these pollutants on the microbial deposition, clearance, and invasion of the test organism are being investigated. Alterations in specific defense systems being studied include the pulmonary macrophage, the cellular immune system, and mucociliary clearance. The dose-response relationship for both ozone and nitrogen dioxide are being investigated to determine if the concentration of the pollutant is more important than the time of exposure in determining the effect on the animal. Such studies are highly important to both short- and long-term standards. Whole animal studies also include alterations in pulmonary function, blood chemistry, specific tissue burdens of inhaled heavy metals and potential carcinogenesis or modification of carcinogens.

A number of model systems are being used to predict effects on man from long-term exposure. These include studies of the uptake and removal of secondary pollutants such as sulfate salts, sulfuric acid, and heavy metals in perfused lungs. The effects of a large number of pollutants on the pulmonary bacterial defense system are being assessed by measurements in vitro of the effect of these pollutants on the death of pulmonary macrophages, release of marker enzymes from the cell, inhibition of the ability of the macrophage to engulf particles (phagocytosis), alterations in the genetic system (DNA and RNA synthesis), metabolic repair (protein synthesis), and morphology. Combination studies of exposure of whole animals and subsequent isolation of organs or cells for study are being undertaken to eliminate complications associated with whole animal studies and to improve the sensitivity of the system. For example, the effects on important recognition systems using the immune system are being studied using the pulmonary macrophage isolated from exposed animals.

The nature of interactions between pollutants and the other factors in the environment were demonstrated by Dr. Gardner. The toxicity of oxidizing pollutants is influenced by the length of exposure to the pollutant, the concentration of the pollutant, the temperature, predisposing disease, other pollutants present (for example, sulfuric acid and ozone), length of time between exposures and physiological state of the animal. Exposure regimens within the program are designed to mimic the cyclic nature of photochemical pollutants observed over major cities. On long-term exposure, continuous and intermittent nitrogen dioxide exposures appear to be equivalent at the same concentration of nitrogen dioxide. Exercise, similar to heavy work, increases pollutant toxicities. Combination exposures of nitrogen dioxide and ozone, similar to the combination exposures of photochemical smog, appear to be additive and not synergistic. On the other hand, sequential exposure to ozone followed by sulfuric acid mists appear to potentiate the toxicity of the combination.

The discussion following Dr. Gardner's presentation centered around the appropriateness of the models currently being used by EPA. It was generally agreed that the program was well founded upon scientifically accepted principles and represented the present state-of-the-art in the measurement of pollutant effects. A need was cited for more detailed information on the lifetime effects of low levels of pollutants. Estimation of "threshold" or no effect levels of pollutants is therefore difficult. Pressure for immediate assessment of pollutant hazards, only short-term fiscal support mechanisms, and limited personnel and exposure facilities have hampered long-term studies.

A need for combination studies of pollutants was clearly illustrated by Dr. Gardner's data. Most studies, because of the shortages cited

above, are single pollutant studies, while the polluted air actually contains large numbers of pollutants mixed in variable ratios. The sulfuric acid-ozone data emphasizes the need for studies of combinations of pollutants.

2. Research Within EPA's Catalyst Research Program (Conference Discussants - Dr. R. E. Lee and Dr. W. Moore): Dr. Lee and Dr. Moore described the research being performed within EPA's Catalyst Research Program. The strategy of this program is to identify automotive emissions, to determine emission factors, and to estimate the consequences of human exposure to these emissions. Participating laboratories include ESRL, which is in charge of emission characterization and atmospheric chemistry; EMSL, which collects and analyzes fuel and also monitors roadside emissions; HERL - Cincinnati, which studies health consequences of whole exhaust; and HERL - Research Triangle Park, which performs inhalation toxicology, human studies, and in vivo and in vitro testing.

Work performed at ESRL has demonstrated that the oxidizing catalyst reduces CO, hydrocarbons, phenols, aldehydes, polynuclear aromatic hydrocarbons, and organic particulate emissions and that there is negligible platinum and palladium emitted from the catalyst. The sulfuric acid emitted is in the ultrafine particle size range (0.01 - 0.10 micron) and sulfuric acid monitoring in this particle size range is difficult to perform. EMSL is presently surveying fuel for sulfur content and has monitored sulfate levels during its L.A. Freeway study. Ongoing studies at HERL - Cincinnati include exposure of rats to metallic sulfate aerosols, manganese compounds such as MMT, and diesel exhaust emissions.

HERL - Research Triangle Park is presently investigating the effects of platinum and palladium, ultrafine sulfuric acid aerosol exposures, and toxicity associated with sulfuric acid and sulfate salts. Clinical studies are now beginning exposing healthy volunteers to 100 $\mu g/m^3$ sulfuric acid aerosols (0.3 microns) for short times.

Discussion brought out the need for a better chemical characterization of the actual atmospheric sulfate particles since the biological effects of sulfate aerosols are highly dependent on the chemical composition of the sulfate salt. The deposition, availability, and biological effect of ultrafine aerosols is unknown since the detection, measurement, and production of these submicron particles have only just evolved as a new technology. The study of these aerosols in animals and further characterization of the toxic effects of sulfate and sulfuric acid aerosols in animals is needed to speed the human exposure experiments with safety to the participants. In turn, these studies are needed for further epidemiological studies of the population as a whole. Since diesel exhaust emissions represent an entirely new area of pollutant toxicity, and since diesel emissions are of small particle size, future studies in animals in this area were thought justified.

C. Future Development, Trends Related to Automotive Power Plants, Fuels, and Control Devices (Conference Discussant - Dr. R. Bradow): Dr. Bradow discussed the future development and trends related to automotive power plants, fuels, and control devices as well as the characterization of primary and secondary pollutants associated with mobile sources. Presently NO_x emissions are decreased by recirculating exhaust gas back to the combustion chambers. In the future, reducing catalysts will be used to convert nitrogen dioxide to nitrogen, but ammonia production could also occur. At first, the solution seemed to be a dual bed configuration for the catalyst, the first being a reducing catalyst to convert NO_x to nitrogen, then an oxidizing bed. If ammonia is produced, however, this second catalyst would oxidize the ammonia to NO_x again. A ruthenium catalyst selectively forms nitrogen, but also forms ruthenium oxide which is extremely volatile. A rhodium reducing catalyst is nonselective so the design would have to incorporate an air flow controller to favor stoichiometrically nitrogen formation. However, to control the air flow the oxygen concentration in the converter must be monitored. Volvo and Saab have fitted zirconium oxide sensors in the 1977 California cars for this purpose. The reliability of oxygen sensors will determine the converter efficiency.

Alternative power plants to the gasoline internal combustion plants are in various stages of development. Diesel combustion engines emit low amounts of hydrocarbons and carbon monoxide, but, due to their low operating temperatures, the use of catalytic converters as an exhaust control device would seem technically difficult. They also emit increased levels of polynuclear aromatic hydrocarbons. Gas turbines release 1 to 1.5 g/mile NO_x and can burn a variety of fuels. The Sterling cycle engine is also insensitive to fuel, but prototypes are nonexistent. An engine that is gaining increasing popularity is the stratified-charged engine, which uses a pre-combustion chamber to allow combustion of lean fuel mixtures.

Secondary products that have been identified in converted auto exhaust include hydrocarbons (HC), ammonia (NH_3), nitrous oxide (N_2O), methylamine, and hydroxylamine. Cobalt and nickel oxides have been detected in exhaust and probably are derived from the alumina support of the three-way catalyst.

Dr. Bradow suggested that there may be major changes in the power plant composition of the automotive fleet in the near future. Diesel power plants, the stratified-charge engine, and power plants less sensitive to fuel requirements will increase due to economic demands for more efficient plants and restrictions on fuel availability through oil conservation.

A considerable discussion followed emphasizing the need for rapid dissemination of the host of technical information being developed by emission scientists. The chemical complexities of exhausts and the differences between raw, converted, and multiple bed converted exhausts make estimation of the health effects of such transitions in technology difficult to predict. The dissimilarities between gasoline internal combustion and diesel exhaust will require added study of the effects of diesel exhausts themselves. The list of chemical compounds found as primary emission seems endless and priorities will have to be determined for health assessment. Resources are insufficient to test each compound individually in either in vitro or in vivo systems. On the other hand, studies of whole exhaust may be equally misleading because interaction between pollutants may mask important effects of a particularly hazardous pollutant present in low concentrations. Dr. Bradow pointed out the need for continued study of converters as they are functioning in the field since maintenance, tampering, and reliability of control devices will have major effects upon the pollutants emitted. A sentiment was expressed for the need for a closer coupling of the development of control technologies and health effects assessment. Workshops such as the present conference were favored as filling the need for rapid distribution of information and interaction between health and physical scientists.

D. Atmospheric Chemistry: Transport and Transformation of Primary and Secondary Pollutants:

1. Conference Discussant - Dr. A. P. Altshuller: Atmospheric chemistry, the transport and transformation of primary and secondary pollutants, was discussed by Dr. Altshuller. Particles are known to grow rapidly and physical growth will alter the properties of primary emissions. Initial exhaust aerosols of sulfuric acid are 0.01 to 0.10 microns in size, but during aging (approximately 20 seconds) grow to 0.2-0.3 microns in size, for example. Ammonia, present in the atmosphere will react with sulfuric acid to form ammonium sulfate. Nitric oxide, present in roadways approaching 1 ppm will react with ozone to form nitrogen dioxide, but the degree to which this occurs has not been quantitated. On the urban scale, the automobile contributes appreciably to the atmospheric hydrocarbon burden. High sulfate and nitrate concentrations are associated with stagnating high pressure anticyclones, which occur with the highest frequency just west of the Appalachians.

Sampling problems exist for nitrates. Chemical alteration on glass filters produces artifacts and makes detection of the chemical species of oxides of nitrogen difficult. Quartz and Teflon filters lead to fewer artifacts. Much of the nitrate in the air is present as nitric acid, but particulate nitrate and peroxyacetyl nitrates (PAN) are present in appreciable amounts, also.

Dr. Altshuller, like Dr. Bradow, emphasized the recent advances in atmospheric chemistry that have led to greater detailed knowledge of the specific chemical species present in the atmosphere. As a consequence, a great list of compounds has been prepared that outstrips present resources for health effects evaluation. Automotive exhaust, while a major contributor to air pollution, becomes mixed and reacts with other pollutants in air from non-automotive sources. As a result, once emitted into the air, automotive exhausts must be considered an integral part of the overall general problem of air pollution and not as a separate entity.

2. (Conference Discussant - Dr. J. Pitts): Dr. Pitts stated that it was his view that one could not pinpoint the most important atmospheric reactions due to transportation-related pollutants without accurate and detailed inventories for a variety of transportation systems in different types of air basins. Furthermore, one can not generate a reliable chemical, physical, and/or biological model capable of predicting the air quality impact of a given type of transportation without detailed emission inventories for all concurrent sources of air pollution. Thus, similar needs for accurate inventories apply to exhaust and evaporative losses, etc., from other transportation systems (aircraft, older cars, diesel engines, etc.) as well as, of course, to emissions from stationary sources.

As an example of the difficulties in establishing such information, knowledge only of the carbon monoxide and non-methane hydrocarbon emissions from light-duty motor vehicles (LDMV) equipped with new catalyst systems is clearly insufficient. It is most important to know what happens to emissions as LDMV catalysts age. The amount and chemical composition of a number of organic oxygenates (alcohols, aldehydes, ketones, acids, and other simple and complex oxygenated nitrogenous compounds, such as polynuclear aromatic hydrocarbons and their oxygenated products) might change as the catalyst ages. Quantitative data are needed. Similar data are needed for the stratified charge engines, diesel, and fuel injection systems.

As a methodological approach, Dr. Pitts advocated the mass balance method. That is to say:

C, H, O, N, S, Pb, Mn, etc. atoms <u>into</u> car (i.e. engine and gas tank)	=	C, H, O, N, S, Pb, Mn, etc. atoms coming <u>out</u> (exhaust, evaporative emissions, etc. or left over as engine or exhaust deposits

Specific atmospheric reactions related to transportation-generated pollutants include reactions of oxides of nitrogen. NO and NO_2 are ubiquitous products of all combustion processes, yet a detailed understanding of the rates and mechanisms of a number of the atmospheric reactions is lacking. The photolysis of nitrous acid (HONO) is a driving force in initiating photochemical smog since it produces hydroxyl (·OH) free radical, a key intermediary in the chemistry of the lower and upper atmosphere. A number of measurements for the equilibrium reaction of NO and NO_2 to form HONO have been reported but the relative importance of homogenous vs. heterogenous pathways has not been determined. The nitrous acid concentration in the air is still undetermined also.

Nitric acid (HNO_3) has been identified recently at the University of California Riverside Laboratory by the use of Fourier transformed infrared spectroscopy. The reactions leading to nitric acid from NO, NO_2, and water are not known. Ammonium nitrate aerosols are also found in the air and may have biological importance. A new species, peroxylnitric acid (HO_2NO_2) has been identified as the dominant product from the reaction of hydroperoxyl radical with nitrogen dioxide.

Finally, in dealing with the oxides and oxyacids of nitrogen, it is important to recognize that they possibly are the atmospheric precursors of a wide spectrum of secondary pollutants which may represent more toxic hazards to man. Other atmospheric reactions of importance include those between irradiated aromatic hydrocarbons and NO_x, homogenous vs. heterogenous reactions of amines in the presence of NO_x and water vapor, and the oxidation of polycyclic aromatic hydrocarbons. Each of these reactions may lead to more potent biologically active compounds than the parent starting materials. In each case there is a clear need for pressure and temperature dependence kinetic data upon which to build mathematical models of specific climatic conditions and regional areas. Such studies form the basis for projection of environmental effects of different strategies and the resulting impact of specific transportation alternatives.

While the approaches to atmospheric chemistry advocated by Drs. Altshuller and Pitts were dissimilar, their conclusions were remarkably similar. Both concluded that knowledge of the chemistry of oxides of nitrogen was imperative to future decisions on the impact of alternative transportation systems. The need for a sustained research effort on the fundamental mechanisms of the formation of more complex products from the primary pollutants such as NO_x was apparent. Since atmospheric chemistry is a rapidly developing field which is uncovering a more complex inventory of air pollutants that, like auto exhaust chemistry, promises to outstrip the resources available for toxicological evaluation, a close interaction with health scientists is imperative. Priorities for toxicological and epidemiological studies can be drawn from such discussions which will reduce duplicative effort and speed the early recognition of potential health hazards.

E. Research Strategies:

1. Toxicological Studies (Conference Discussant - Dr. D. L. Coffin): The research strategies for toxicological studies in animals and man were discussed by Dr. Coffin. It is important to distinguish between human and animal experiments as it is not always possible to provide direct experimental evidence for toxicity in man. For studies of air pollutant toxicity to occur, a detailed chemical identification of the air pollutant must first have been made and the air pollutant produced for exposure systems in a chemically and physically similar manner to that in air. To date, studies have been restricted to gases, vapors, and conventional aerosols of pure single pollutants or a very few combinations of these pollutants. The question of toxicity is extremely complex and there are no simplistic answers now, or in the future, likely to become available. In particular, interactions between pollutants are very difficult to predict.

Clinical studies in human volunteers are restricted to the very lowest concentrations of pollutants likely to provoke a response. Only the simplest non-invasive methods are ethically possible. Pulmonary mechanical changes are most commonly used and may be inadequate for measurement of subtle effects. Ozone inhalation results in changes in pulmonary function, as does SO_x particulate and NO_2. There is, however, no clear relationship between pulmonary mechanical changes in man with short-term ozone exposure and obstructive lung disease which occurs in animals chronically exposed to ozone. Chronic obstructive lung disease in man, however, appears to be associated with ozone pollution by epidemiological studies. The advantages of human studies are obvious in that there is no need to extrapolate the results to man from another species, no need to assure that the species selected is unduly sensitive or that basic physiological difference might exist. These needs must be dealt with in all animal experiments.

Specific toxicological models applicable to air pollutant studies include infectivity studies which are sensitive to near ambient levels of nitrogen dioxide and ozone, but relatively insensitive to metals and sulfate particulates. Bacterial pneumonias in man are not prevalent and, therefore, these models may be more representative of the role of air pollutants in obstructive bronchitis.

Long-term, low-level exposures of rats and mice in the absence of airborne pathogens are being used to study the development of emphysema. There is a need to translate these observations of pathological changes resulting in emphysema into mechanistic ones, such that early indicators applicable to human studies can be discovered. This is especially true for studies of interaction such as ozone and sulfuric acid mists. Biochemistry and pharmacology are the major tools for these investigations.

A major concern is the mutagenic and carcinogenic properties of vapor phase organic compounds. Using bacterial mutagenic test systems, such as the Ames Salmonella test, either directly or indirectly, through an intermediary host, such as a mouse, mutagenic activity has been detected in the air. As referred to previously, a host of complex organic compounds are present in auto exhaust and in air as secondary products from auto exhaust. These systems are reliable as first screens for positive compounds and for screens to aid in purification of mutagens, but negative results provide little assurance of safety in man. Co-carcinogenesis or promotors present as air pollutants are a major potential problem. There is a 20% excess in the incidence of cancer in man due to urban factors other than smoking and these may include air pollutants among the possible factors.

The immune system has been shown to be depressed by nitrogen dioxide. The uptake of biogenic amines by the lung and the formation of prostaglandin hormones by the lung have also been shown to be depressed by both ozone and nitrogen dioxide. An important variable is the diet of man and animals which has been shown to increase the sensitivity to oxidizing air pollutants during vitamin E deficiency.

2. Epidemiological Studies (Conference Discussant - Dr. R. Horton): Epidemiological studies, like toxicological studies, are dependent upon the unequivocal identification of the air pollutant and the precise determination of the level of the pollutant to which a given population is exposed. To this end, carbon monoxide is an example of a transportation-related air pollutant for which exposure data has been satisfactorily obtained. As a gaseous pollutant, distributive model systems have been developed which predict with accuracy the carbon monoxide exposure of the population. Particulate pollutants such as lead are less well studied. Large particles of lead settle out of the atmosphere and are difficult to sample. Within the respirable particle size range, a close correlation exists between the blood lead level and the atmospheric lead level. Epidemiologists must provide for adequate sampling to estimate the atmospheric level of a pollutant and the pollutant exposure of the population. Occupational and personal habits tend to cause unequal exposures of specific individuals within the population; thus stratification of sampling must be undertaken to correct for unusually high exposures. For example, workers in and around highway tunnels are exposed to higher lead levels than the general population. Taxi drivers may be exposed to levels as high as 9 $\mu g/m^3$. Selection of specific subpopulations may lead to the selection of homogenous population with regard to their exposure history. When further studies are undertaken on other substances originating from transportation sources it would be useful for the investigators to review work done on carbon monoxide and lead from a methodological point of view. Some methods were useful, some were not.

To cope with these difficulties, specialized air monitoring systems sited according to the nature of the epidemiological study, rather than meteorological considerations, may be necessary to determine the pollutant exposure history of that specialized sub-population. Methods of assessment of toxicity are generally restricted to non-invasive methods as discussed above for clinical studies or statistical evaluations of excess mortalities.

Inference from an association between exposure and the measured effect in the population under study depends upon the establishment of a cause and effect relationship in experimental animals. Epidemiological studies are an outgrowth of the more basic studies in experimental animals and clinical studies. Since trends such as excess mortality are demonstrable only over long time periods and since populations must be followed over several climatic cycles, epidemiological studies require a sustained commitment and are expensive. The effort involved in epidemiological studies, and especially in monitoring systems designed to estimate exposures, suggests that baseline studies of air pollutant levels should be maintained whenever feasible. Some markers such as acetylene used as a measure of vapor phase auto emission exposure are disappearing because of the abolition of ethylene from auto exhausts by the catalytic converter. Overlapping data between old and new monitoring systems is needed.

F. Specific Problem Areas:

1. Hydrocarbons (Straight Chain and Aromatics) (Conference Discussant - Dr. R. Suskind): Discussion of specific problem areas began with a consideration of atmospheric hydrocarbons by Dr. Suskind. The characteristics and effects of the hydrocarbons emitted pre-catalyst are well documented. More research is required on the characterization and health effects of the hydrocarbons emitted from diesel engines and the catalytic converter. Combination studies are lacking with regard to vapor phase and particulate hydrocarbons.

Polynuclear aromatic hydrocarbons and their oxidative products, including peroxides, hydroperoxides and endoperoxides, need to be scrutinized in biological systems for possible toxicity. Possible teratogenicity could be studied using mouse limbs in culture. The effects of organics on biogenic amine levels *in vivo* may be an early marker of toxicity.

It must be recognized that time is necessary to perform quality studies. For these to occur, long-term commitments from DOT and EPA are vital.

2. Complex Organics (S, N, O, and Other) (Conference Discussant - Dr. J. Rowlands): While many complex organic compounds have been identified in the atmosphere from air pollution, little, if any, information on the toxicity of these compounds is available. An example is peroxyacetylnitrate (PAN) which has been identified as the major cause of lacrimation in photochemical smog. PAN is also a potent oxidizing compound, but no long-term low-level studies of PAN's effects on the lung or other mammalian systems have been undertaken. The potential importance of these compounds in mutagenesis and carcinogenesis suggests that this area is in need of review and reorientation in priorities. Since little hard data on biological effects exists, substantive recommendations can not be made at this time.

3. Particulate, Especially Tire Dust (Conference Discussant - Dr. A. Zarkower): Atmospheric particulates as a transportation-related pollutant, especially tire dust, were discussed by Dr. Zarkower. The components of tire dust include stearic acid, zinc oxides, antioxidants, paraffinic waxes, alkyl tin compounds, and possible cadmium. Extender oils account for 20% of tire rubber. They are highly volatile and benzo(a)pyrene is thought to be present. Examples of other rubber additives are: organic sulfur compounds, such as thiram, a skin sensitizer; carbon black, a possible source of benzo(a)pyrene; volcanizer amines; and carbamates, potent pharmacological agents.

Roadside particulate levels can be as high as 160 $\mu g/m^3$. The major constituents are: carbonaceous (87.5 $\mu g/m^3$), quartz (23.1 $\mu g/m^3$), limestone (34.6 $\mu g/m^3$), oil soot (3 $\mu g/m^3$), iron oxide (3 $\mu g/m^3$), tire wear (8 $\mu g/m^3$), and selenium (3 $\mu g/m^3$). The carbonaceous component associated with the tire is not respirable, and is in that way similar to the other roadside particulate.

Health effects due to the compounding of rubber are well documented. Thirty percent of rubber workers suffer from various forms of contact dermatitis. There is an increased trend toward bladder cancer in rubber workers. Recycled tires used in shoes lead to an increased incidence of dermatitis. The active agents might be the thiurams, diphenyl guanidine or the mono benzyl ether of hydroquinone.

While there are definitive adverse health effects associated with tire and rubber manufacture, tire dust does not appear to be a major transportation-related health hazard since little of the degradation product appears as respirable dust. As a consequence, Dr. Zarkower presented his views on additional research strategies for approaches to the general problem of airborne particulates as primary and secondary air pollutants.

Research strategies in regard to particulates which were recommended by Dr. Zarkower include:

(1) Inhalation toxicology using whole animals

  (A) Acute studies at high levels to demonstrate possible effects

  (B) Chronic exposures at ambient levels

  (C) Study alveolar macrophage inactivation and lung fibrosis

  (D) Gastrointestinal effects due to swallowing

(2) Reticuloendothelial system

(3) In vitro alveolar macrophage studies

In the discussion it was generally agreed that tire degradation products are substantial contributors to transportation-related pollution, but because of the large particle size dominating, tire degradation products have little direct impact on human health. The workshop agreed that it would be prudent to review the subject at periodic intervals to ensure that technological alterations in tire production or use did not alter future trends. (Editorial Note: Subsequent to the workshop R. L. Williams and S. H. Cadle (Characterization of Tire Emissions Using an Indoor Test Facility, G. M. Research Laboratories Publication GMR-2483, 1977) reported little production of respirable particles during tire degradation. Straight-ahead, high-speed driving resulted in the smallest particle distribution and highest tire degradation rate, yet these conditions are not likely to present a significant respirable particle burden. The observation by these authors of measurable amounts of extender oil vaporization and some technical limits on detection of particle size distribution verifies the workshop's conclusion that continued surveillance is needed.)

The discussion of Dr. Zarkower's general remarks concerning approaches to the assessment of respirable particles concluded that studies of the reticuloendothelial system might merit more detailed consideration. This conclusion is covered in the general recommendations of the workshop. Discussion was provoked over the aerosols generated by aircraft, especially jet aircraft. Little hard data were brought up to either substantiate or negate a potential role for jet engine emissions as a source of either secondary or primary particulate emissions. The role of aircraft can not be assessed at this time.

4. NO_x and other Automotive Emissions (Conference Discussant - R. Ehrlich): Dr. Ehrlich presented data on the use of the infectivity model for the detection of toxicity of low concentrations of nitrogen dioxide, ozone and several sulfates. This animal model system, which measures the effect of inhalation of air pollutants on the resistance to bacterial or viral respiratory infections, appears to be highly sensitive and reproducible. The parameters used to assess the health effects include, mortality and survival rates, retention of inhaled bacteria in the lungs, conventional and scanning electron microscopy of the respiratory tract, activity of alveolar macrophages and a variety of immunological responses. Details of experimental results were reported indicating an additive effect of a single short-term (3 hr) exposure to low concentrations of nitrogen dioxide and ozone mixtures, and an apparent synergistic effect upon repeated daily acute exposures to mixtures of the same pollutants. Additional information was presented on the effects of short-term exposures to zinc sulfate, zinc ammonium sulfate and ammonium sulfate alone, in combinations, or in presence of other pollutants such as nitrogen dioxide and ozone on the resistance to bacterial pneumonia. The impairment of defense against respiratory infections by exposure to the various pollutants underscores Dr. Gardner's suggestion that the primary site of injury may be the pulmonary macrophage.

During the discussion, points were raised to support the extended use of morphological and biochemical studies in the infective model. There was general agreement that both types of studies are needed to draw better clinical correlates with human disease. This is particularly true in the case of heavy metals and sulfate particulates. Failure to detect injury by the infectivity model does not mean that there is no human effect. Much of the work to date has been limited to the very pressing problem of setting standards and criteria. Underlying principles leading to the disease state have been studied only to a limited extent and general principles have not been drawn to provide a guide for future toxicants. A general conclusion of the workshop was a need for investigations of mechanism of action of pollutants. Classical mortality studies, likewise, were considered essential. A real need was felt for studies of ultrafine aerosols of sulfuric acid and sulfate alone or in mixtures with other pollutants such as nitrogen dioxide, heavy metals, and carcinogens. Such work depends upon the availability of an ultrafine aerosol generating and sampling facility. EPA plans to have one such exposure system operating during 1977, but the volume of work will exceed the available time on this chamber. An additional facility is needed.

Table 1

Summary of Transportation-Related Pollutant Research
at EPA Using Whole Animal Systems*

ANIMAL, IN VIVO					
POLLUTANT	PULMONARY FUNCTION	SERUM CHEMISTRY	TISSUE BURDENS DEPOSITION/CLEARANCE		CARCINO-GENESIS
			LUNG	OTHER	
NO_2		C c c			
$NO_2 + O_3$					
H_2SO_4	C e a,c	C i a			C e c
$H_2SO_4 + O_3$					
H_2SO_4 + PARTIC-ULATES					
SO_2	C e a		C e a		
SULFATES	C e a		C e c		
COMPLEX MIXTURE	C i a				
Mn SALTS					
MnO_X			F e a		
Pt SALTS			C e a		
PtO_X					C e c
Pd SALTS			C e a		
PdO_X					
Pb SALTS					
PbO_X					C e c
BENZO(a) PYRENE					C e c

*i = in house
e = extramural

a = acute (< 1 day)
c = chronic

C = Current
F = Future
P = Past

Table 2

Summary of Transportation-Related Pollutant Research at EPA Using Whole Animal Systems

	ANIMAL, IN VIVO								
	MICROBIAL INTERACTION				SPECIFIC DEFENSE SYSTEMS				PATHO-HISTOLOGY
						Pulmonary	Immune System		
Pollutant	Infectivity	Deposition	Pulmonary Clearance	Invasion of Blood	Alveolar Macrophage	Free Cell Populations	Humoral	Cell Mediated	Ciliary Function	Lung
NO_2	C i a,c						P i,e c	P e c		
$NO_2 + O_3$	C e a,c	C e a,c	C e a,c							
H_2SO_4	C i a	P i a	P i a		C e a,c		C i a	C i a	C i,e a	C e a,c
$H_2SO_4 + O_3$	C i a	F i a	F i a	F i a			F e a	F e a	F i a	
H_2SO_4 + Particulates	C e a,c		C e a,c				C e c			C e a,c
SO_2	C e c	F e c	F e c		C e a,c					
Sulfates	C e a		P i a		C e a,c					
Complex Mixture	C i a		C i a,c							
Mn Salts									C e a,c	
MnO_x	C e a	C e a	C e a	F e a	F e a	F e a	C e a		C e a,c	
Pt Salts					P e c	P e c		C e a		
PtO_x	C e a	C e a	C e a		C e a					C e a,c
Pd Salts					P e c	P e c		C e a		
PdO_x	C e a	C e a	C e a		C e a					C e a
Pb Salts						P e c				
PbO_x						P e c				C e c
Benzo(a) Pyrene										C e c
CO										

Table 3

Summary of Transportation-Related Pollutant Research at EPA Using In Vitro Testing Systems

	IN VITRO														
	CYTOTOXICITY		Phagocytosis (AM)	Enzymes			Uptake of Precursors for Synthesis of								
							DNA		RNA		PROTEIN		Morphology		
Pollutant	AM	WI-38	ATP/cell	AM	WI-38	Other	WI-38	Other	WI-38	Other	WI-38	Other	(AM)	Mutagenesis	Neoplastic Transformation
Sulfates, Sulfites	i C a		i C a	i C a									i C a		
Mn Salts	i P a	i P a	i P a	i P a	i P a		i P a		i P a		i P a		e P a	i F a	i F a
MnO_x	i P a														i F a
Pt Salts	i P a	i P a	i P a	i P a	i P a		i P a		i P a		i P a		i P a	e C a	i F a
PtO_x	i P a	i P a	i P a	i P a	i P a		i P a		i P a		i P a		i P a		i F a
Pd Salts														i F a	i F a
PdO_x	i P a												i P a		
Pb Salts															
PbO_x	e C a		e C a	e C a									e C a		
SO_2	e C a	e C a	e C a	e C a			e C a		e C a						
Diesel-engine Exhaust	e C a	e C a	e C a	a	e C a	e C a								e C a	
Ni Salts	i C a	i P a	i P a	i P a	i P a		i P a		i P a		i P a		i P a	i F a	i F a
NiO	i C a														

III. AGENDA

Wednesday, June 15

9:00 - 9:30	1.	Objectives of DOT	Mr. Samuel Coroniti DOT, Washington, D.C.
9:30 - 11:30	2.	Overview of Ongoing Research on Automotive-Related Pollutants	
		A. Overall	Dr. Donald Gardner EPA, RTP
		Break	
		B. Within EPA's Catalyst Research Program	Dr. Robert Lee EPA, RTP Dr. Wellington Moore EPA, Cincinnati
11:30 - 12:00	3.	Summary	Dr. Gordon Hueter EPA, RTP
12:00 - 1:00	Lunch		
			Discussion Leader
1:00 - 1:45	4.	Future Development, Trends Related to Automotive Power Plants, Fuels, and Control Devices	Dr. Ronald Bradow EPA, RTP
1:45 - 2:30	5.	Characterization of Primary and Secondary Pollutants Associated with Mobile Sources	Dr. Ronald Bradow EPA, RTP
2:30 - 3:00	Break		
3:00 - 4:30	6.	Atmospheric Chemistry: Transport and Transformation of Primary and Secondary Pollutants	Dr. A. Paul Altshuller EPA, RTP Dr. James Pitts Univ. of California at Riverside
4:30 - 5:00	7.	Proposed Charges of Conference	Dr. Gordon Hueter EPA, RTP
5:30 - 6:30	Social Hour		
6:30 - 7:30	Dinner		
7:30 -	Informal Discussion Period		

Thursday, June 16

		Discussion Leader
9:00 - 9:15	1. Review of Charges	Dr. Gordon Hueter EPA, RTP
	2. Research Strategy	
	A. Toxicology	Dr. David Coffin EPA, RTP
10:15 - 10:45	B. Epidemiology	Dr. Robert Horton EPA, RTP
10:45 - 11:00	Break	
	3. Specific Problem Areas	
11:00 - 11:30	A. Hydrocarbons (straight-chain and aromatics)	Dr. Ray Suskind Univ. of Cincinnati
11:30 - 12:00	B. Complex Organics (S, N, O, and Other)	Dr. John Rowlands Southwest Foundation for Res. and Education
12:00 - 1:00	Lunch	
1:00 - 1:30	C. Particulates (especially tire dust)	Dr. Arian Zarkower Penn. State Univ.
1:30 - 2:00	D. NO_x and Other Automotive Emissions	Dr. Richard Ehrlich IIT Research Inst.
2:00 - 2:15	Break	
2:15 - 3:00	4. Priorities and Procedures	Dr. Gordon Hueter EPA, RTP
3:00 - 4:00	5. Recommendations (discussion and adoption)	Dr. Daniel Menzel Duke Univ.

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